Co$_3$V$_2$O$_8$ is an insulating system with Co$^2+$ spins (fictitious spin $S=1/2$) arranged in a kagome staircase structure or in a buckled kagome plane. It is interesting to investigate the magnetic behavior of this two-dimensional spin system at low temperature $T$, because it has the geometrical frustration inherent in its crystal structure. Another interesting viewpoint of this system is their dielectric properties, because for the iso-structural system Ni$_3$V$_2$O$_8$, the multiferroic behavior has been reported, that is, magnetic and ferroelectric transitions take simultaneously at a critical temperature.[1] Theoretically, the mechanisms of the multiferroic behavior are proposed to be expected in system with the transverse spiral order.[2] The magnetic structure of Co$_3$V$_2$O$_8$ has also been investigated by neutron diffraction study. The detailed results and discussions are reported in ref. 3.

We have carried out the magnetization, specific-heat and dielectric measurements on single crystal samples Co$_3$V$_2$O$_8$ in various magnetic fields along three crystallographic axes up to 5 T. Neutron measurements on a powder sample as well as a single crystal with magnetic field along $c$ have been carried out down to 2 K. Neutron measurements on a single crystal were carried out at T1-1 (HQR), where the double axis condition was adopted. In zero magnetic field, the system exhibits three transitions at temperatures $T_1 \approx 11.2$ K, $T_2 \approx 8.8$ K and $T_3 \approx (6.0-7.0)$ K, where the last transition has the co-existence region of $T$ due to its first order nature. No evidence for ferroelectric transitions has been observed in the measurements of the dielectric constant with the electric fields along three crystallographic axes, $a$, $b$ and $c$.

On the basis of the results of the neutron scattering study, detailed magnetic structures are proposed in zero magnetic field. The ordered moments have the sinusoidal nature with the modulation vector $(0,\Delta,0)$ in the antiferromagnetic phases ($T_3<T<T_1$), and the single crystal data present clear evidence for the non-collinear nature of the magnetic structures in all magnetically ordered phases. The transverse spiral state does not exist in Co$_3$V$_2$O$_8$, which is in a clear contrast with the case of the multiferroic Ni$_3$V$_3$O$_8$. It is consistent with the fact that no evidence for the occurrence of the ferroelectric transition has been found in the dielectric data, that is, the difference between Co$_3$V$_2$O$_8$ and Ni$_3$V$_3$O$_8$ can be understood by the existing theories. Only small dielectric anomalies closely connected with the magnetic phase transitions have been found.

We have constructed detailed field ($H$)-temperature ($T$) phase diagrams for the fields parallel to the three crystallographic directions $a$, $b$ and $c$ as shown in the figure. The abbreviations P, ICAF, CAF and CF represent paramagnetic, incommensurate-antiferromagnetic, commensurate-antiferromagnetic and commensurate-ferromagnetic phases, respectively. The delta value is estimated from the position of the 02-$\Delta$0 magnetic reflections. Solid symbols are the transition temperatures determined by the magnetization and specific-heat data. The phase boundaries plotted by the open symbols are determined by the neutron scattering studies in the magnetic field $H(//c)$. The dotted lines are drawn to connect the crosses defined as the inflexion points of the M-T curves, and may not correspond to the phase boundaries. In the $T$-region below these inflexion points, the relatively large ferromagnetic component is induced by the applied magnetic field. The
phases stabilized by the external field are very sensitive to the strength of the field. In the magnetic field $H$ along $c$, the magnetically ordered phases with $\delta = 1$ and $\delta = 2/3$ have been found in the $H$-region of $H > 1$ T. The existence of various phases can be considered to be due to the geometrical frustration.

References

Fig. 1. The $H$-$T$ phase diagrams of Co$_3$V$_2$O$_8$ for the magnetic fields $H$ along (a)H//a, (b)H//b and (c)H//c.